

**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re the Application of

Shigekazu TERANISHI et al.

Group Art Unit: 1794

Application No.: 10/578,666

Examiner: M. NELSON

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Docket No.: 126497

For: HEAT-RESISTANT LUBRICITY IMPARTING COATING AGENT, AND  
THERMAL TRANSFER RECORDING MEDIUM

**DECLARATION UNDER 37 C.F.R. §1.132**

I, IZUMI SHINYA, a citizen of Japan, hereby declare and state:

1. I have a Master of Engineering degree which was conferred upon me by The University of Shiga Prefecture, Engineering Graduate School in Hikone-shi, Shiga, Japan in March, 2002.

2. I have been employed by Natoco Co., Ltd. since April, 2002 and I have had a total of 7 years of work and research experience in the field of coating materials.

3. I have a professional relationship with the Assignee of the above-identified patent application. In the course of that professional relationship, I received compensation directly from the Assignee for my work relating to the experiments described herein. I am being compensated for my work in connection with this Declaration.

4. I and/or those under my direct supervision and control have conducted the following tests:

Purposes of Experiment:

To confirm that, in a heat-resistant lubricity imparting coating agent containing a polydimethylsiloxane copolymer including an alkyl group, if the polydimethylsiloxane copolymer contains a fluorinated alkyl group as the alkyl group, it has poor appearance, film forming properties, and compatibility as compared with a polydimethylsiloxane copolymer containing no fluorinated alkyl group.

Experimental details:

In an experiment, resin solutions (Resin solutions #1 to #4) each containing a polydimethylsiloxane copolymer including a fluorinated alkyl group and a resin solution (Resin solution #5) containing a polydimethylsiloxane copolymer including an alkyl group (but including no fluorinated alkyl group) were produced and examined regarding appearance, film forming properties, and compatibility.

**I. Production of Resin solutions:**

(i) Resin solution 1:

An agitator, a thermometer, a condenser, a nitrogen supply pipe, and a 500-ml flask with a monomer dropping tank were prepared. In this flask, 200 parts by weight of isopropyl alcohol, 25 parts by weight of light acrylate FA-108 (KYOEISHA CHEMICAL CO., LTD.; chemical name: perfluorooctyl ethyl acrylate), and 25 parts by weight of FM-0711 (CHISSO CORPORATION; product name: polydimethylsiloxane containing a methacrylic group at one end, having a silicone chain length of 1000) were heated to 50°C under nitrogen atmosphere to prepare a reaction system.

Subsequently, 3 parts by weight of lauryl mercaptan and 1.5 parts by weight of AIBN (WAKO KAGAKU INDUSTRY CORPORATION; chemical name: 2,2-

azobisisobutyronitrile) were added and heated to 75°C for 30 minutes and then polymerized for 7 hours at 75°C. In this way, Resin solution 1 containing a polydimethylsiloxane copolymer including a "fluorinated alkyl group" was produced.

The light acrylate FA-108 (KYOEISHA CHEMICAL CO., LTD.; chemical name: perfluorooctyl ethyl acrylate) corresponds to an ethylenically unsaturated monomer containing a fluorinated alkyl group (A-1) disclosed in Kazuyoshi (JP-A-05-185757).

This Resin solution 1 corresponds to Embodiment 5 of Kazuyoshi.

(ii) Resin solution 2:

For comparison with Resin solution 1, Resin solution 2 was produced in the same manner as the method of Resin solution 1 except replacing isopropyl alcohol with methyl ethyl ketone. In this way, Resin solution 2 containing a polydimethylsiloxane copolymer including a "fluorinated alkyl group" was produced.

This Resin solution 2 corresponds to Embodiment 5 of Kazuyoshi.

(iii) Resin solution 3:

For comparison with Resin solution 1, Resin solution 3 was produced in the same manner as the method of Resin solution 1 except changing the dosage of light acrylate FA-108 to 12.5 parts by weight, changing the dosage of FM-0711 to 22.5 parts by weight and adding 15 parts by weight of lauryl acrylate. In this way, Resin solution 3 containing a polydimethylsiloxane copolymer including a "fluorinated alkyl group" was produced.

This Resin solution 3 corresponds to Embodiment 6 of Kazuyoshi.

(iv) Resin solution 4:

For comparison with Resin solution 1, Resin solution 4 was produced in the same manner as the method of Resin solution 1 except changing the dosage of light acrylate FA-108 to 12.5 parts by weight, changing the dosage of FM-0711 to 5 parts by weight and adding 15 parts by weight of lauryl acrylate, 2.5 parts by weight of FM-7721 (CHISSO

CORPORATION; product name: polydimethylsiloxane containing a methacrylic group at both ends, having a silicone chain length of 5000), and 15 parts by weight of BLEMME PSE-1300 (NOF CORPORATION, chemical name: stearyoxy polyethylene glycol monomethacrylate). In this way, Resin solution 4 containing a polydimethylsiloxane copolymer including a "fluorinated alkyl group" was produced.

This Resin solution 4 corresponds to Embodiment 6 of Kazuyoshi.

(v) Resin solution 5:

Identical devices to those in the method of producing Resin solution 1 were used. Put in a flask were 100 parts by weight of toluene, 100 parts by weight of methyl ethyl ketone, 30 parts by weight of SMA (stearyl methacrylate), 35 parts by weight of MMA (methyl methacrylate), 12.5 parts by weight of HEMA (hydroxyethyl methacrylate), 12.5 parts by weight of methacrylic acid, and 10 parts by weight of VPS 1001 (WAKO PURE CHEMICAL INDUSTRIES, LTD., product name: polydimethylsiloxane amide containing an azo group, having a silicone chain length of 10000).

This mixture was agitated to be uniformized and then heated to 80°C and polymerized for 5 hours. Thereafter, 0.5 parts by weight of ABN-E (JAPAN HYDRAZINE COMPANY, INC., product name: 2,2'-azobis(2-methylbutyronitrile)) was dropped into the mixture, which was then polymerized for 2 hours. In this way, Resin solution 5 containing a polydimethylsiloxane copolymer including a long chain alkyl group having a carbon number of 18 (but including no fluorinated alkyl group) was produced.

It is to be noted that stearyl methacrylate corresponds to a vinyl monomer containing a long chain alkyl group having a carbon number of 18 (an alkyl group constituted of only a carbon element and a hydrogen element). Resin solution 5 corresponds to Resin solution 12 in Embodiment 12 of the present application (see Fig. 3).

Table 1, below, shows the compositions of Resin solutions 1-5.

Table 1

	Resin solution 1	Resin solution 2	Resin solution 3	Resin solution 4	Resin solution 5
FA-108	25	25	12.5	12.5	
FM-0711	25	25	12.5	5	
VPS1001					10
FM-7721				2.5	
PSE-1300				15	
Lauryl acrylate			15	15	
MMA					35
SMA					30
HEMA					12.5
Methacrylic acid					12.5
AIBN	1.5	1.5	1.5	1.5	
ABN-E					0.5
Lauryl mercaptan	3	3	3	3	
Isopropyl alcohol	200		200	200	
Methyl ethyl ketone		200			100
Toluene					100

**II. Appearance of Resin solutions:**

(i) The above Resin solutions 1 to 5 were evaluated regarding the appearance by visual observation. The results of this evaluation are shown in Table 2.

Table 2

	Resin solution 1	Resin solution 2	Resin solution 3	Resin solution 4	Resin solution 5
Appearance of Resin solution	X	X	Clear and Colorless	Clear and Colorless	Clear and Opaque white

X: Turbidity and Precipitation were observed

(ii) As shown in Table 2, turbidity and precipitation were observed in Resin solutions 1 and 2 (corresponding to Embodiment 5 of Kazuyoshi). This seems because the polydimethylsiloxane copolymer in Resin solutions 1 and 2 contains a "fluorinated alkyl group."

Specifically, a fluorinated component is low in solubility in a solvent and therefore the copolymer containing the fluorinated component is also conceived to be low in solubility in a solvent. Accordingly, turbidity and precipitation were observed in Resin solutions 1 and 2.

(iii) On the other hand, neither turbidity nor precipitation was observed in Resin solution 5 (corresponding to Resin solution 12 in Embodiment 12 of the present application). This seems because the polydimethylsiloxane copolymer in Resin solution 5 contains no "fluorinated alkyl group" and hence it has a good solubility in a solvent.

### III. Film forming property:

(i) Resin solutions 1 to 5 were evaluated regarding film forming properties. Specifically, 50 parts by weight of each of Resin solutions 1 to 5 was added with a mixed solvent of 45 parts by weight of methyl ethyl ketone and 5 parts by weight of cyclohexanone. These mixtures were separately coated on the surfaces of PET films (TOYOBO CO., LTD, Cosmo Shine A4300) of base materials so that the thickness of each coating film after a drying process is about 1  $\mu\text{m}$ . After letting it stand for 24 hours at room temperature, each coating film was observed. The results thereof are shown in Table 3.

Table 3

	Resin solution 1	Resin solution 2	Resin solution 3	Resin solution 4	Resin solution 5
Repel of coating film	Many observed	Many observed	Not observed	Not observed	Not observed
Haze of coating film	Observed	Observed	Not observed	Not observed	Not observed
Tack of coating film	Observed	Observed	Observed	Observed	Not observed

In Table 3, the term "Repel" represents that part of a coating film does not stick to a PET film, the term "Haze" represents that part of a coating film is hazy, and the term "Tack" represents that the surface of a coating film is sticky.

(ii) As shown in Table 3, many repelling portions ("Repel") were observed in the coating films produced by use of Resin solutions 1 and 2 (corresponding to Embodiment 5 in Kazuyoshi). This is believed to be because the polydimethylsiloxane copolymer contained in Resin solutions 1 and 2 includes a "fluorinated alkyl group" and hence it is hard to stick to the PET film.

In the coating films made by using Resin solutions 1 and 2, "Haze" and "Tack" were also observed. This is believed to be because the ethylenically unsaturated monomer containing a fluorinated alkyl group (in the present experiment, light acrylate FA-108) has low reactivity and thus a number of un-reacted monomers remain in Resin solutions 1 and 2. In Resin solutions 3 and 4 (corresponding to Embodiment 6 in Kazuyoshi), Tack was observed.

Thus, the coating films produced by using Resin solutions 1 to 4 may cause adhesion of dust, ink, etc. to the film surface, leading to printing failures. The coating films produced by using Resin solutions 1 to 4 correspond to a synthetic resin layer of a thermal transfer material of Kazuyoshi.

(iii) On the other hand, any of "Repel", "Haze", and "Tack" were not observed in the coating film produced by using Resin solution 5 (corresponding to Resin solution 12 in Embodiment 12 of the present application). The reason no "Repel" was found is believed to be because the polydimethylsiloxane copolymer contained in Resin solution 5 does not include a "fluorinated alkyl group." The reason why no "Haze" and "Tack" were observed is believed to be because Resin solution 5, unlike Resin solutions 1 to 4, does not contain the low-reactive ethylenically unsaturated monomer containing a fluorinated alkyl group (in the present experiment, light acrylate FA-108) and thus less un-reacted monomers remain in Resin solution 5. The coating film produced by using Resin solution 5 corresponds

to a heat-resistant lubricious protective layer of a thermal transfer recording medium in claim 25 of the present application.

**IV. Compatibility:**

- (i) Resin solutions 1 to 5 were evaluated regarding compatibility.

Specifically, each of Resin solutions 1 to 5 was mixed with a resin (polyvinyl acetal or soluble cotton). These mixtures (1 to 10) were visually observed. The results thereof are shown below in Table 4. Mixtures 1 to 10 were produced in the following manner.

- (ii) Mixtures 1 to 5:

5 parts by weight of polyvinyl acetal was dissolved in 10 parts by weight of MEK (methyl ethyl ketone) and 10 parts by weight of cyclohexanone. This mixture was further mixed with 75 parts by weight of Resin solution 1 to 5 separately and agitated, thereby producing Mixtures 1 to 5.

- (iii) Mixtures 6 to 10:

5 parts by weight of soluble cotton was dissolved in 2.14 parts by weight of IPA (isopropyl alcohol), 8.93 parts by weight of MEK (methyl ethyl ketone), and 8.93 parts by weight of toluene. This mixture was further mixed with 75 parts by weight of Resin solution 1 to 5 separately and agitated, thereby producing Mixtures 6 to 10.



Table 4

	M1	M2	M3	M4	M5	M6	M7	M8	M9	M10
Polyvinyl acetal	5	5	5	5	5					
Soluble cotton						5	5	5	5	5
IPA						2.14	2.14	2.14	2.14	2.14
MEK	10	10	10	10	10	8.93	8.93	8.93	8.93	8.93
Cyclohexanone	10	10	10	10	10					
Toluene						8.93	8.93	8.93	8.93	8.93
Resin solution 1	75					75				
Resin solution 2		75					75			
Resin solution 3			75					75		
Resin solution 4				75					75	
Resin solution 5					75					75
Status of Mixture	X	Y	Y	Y	Z	X	Y	Y	Y	Z

M: Mixture; X: Turbidity and Precipitation were observed; Y: Precipitation was observed;  
Z: Clear and Opaque white

(iv) As shown in Table 4, turbidity and precipitation were observed in Mixtures 1 and 6 (corresponding to Embodiment 5 in Kazuyoshi). Precipitation was observed in Mixtures 2 and 7 (corresponding to Embodiment 5 in Kazuyoshi) and Mixtures 3, 4, 8, and 9 (corresponding to Embodiment 6 in Kazuyoshi). Specifically, Resin solutions 1 to 4 containing the polydimethylsiloxane copolymer including a “fluorinated alkyl group” were poor in compatibility with other resins (in the present experiment, polyvinyl acetal or soluble cotton).

On the other hand, neither turbidity nor precipitation was observed in Mixtures 5 and 10 (corresponding to Embodiment 20 of the present application). Specifically, Resin solution 5 containing the polydimethylsiloxane copolymer including no “fluorinated alkyl group” was good in compatibility with other resins (in the present experiment, polyvinyl acetal or soluble cotton).

#### Conclusions:

The coating agent containing the polydimethylsiloxane copolymer including a “fluorinated alkyl group” was not good in any characteristics, i.e., appearance, film forming

property, and compatibility. On the other hand, the coating agent containing the polydimethylsiloxane copolymer including no "fluorinated alkyl group" (i.e., including another alkyl group) was good in every characteristic, i.e., appearance, film forming property, and compatibility.

In conclusion, exclusion of the fluorinated alkyl group from an alkyl group to be contained in the polydimethylsiloxane copolymer can improve appearance, film forming property, and compatibility of a coating agent.

5. I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date: 2009. 7. 15

Shinya Izumi  
IZUMI SHINYA